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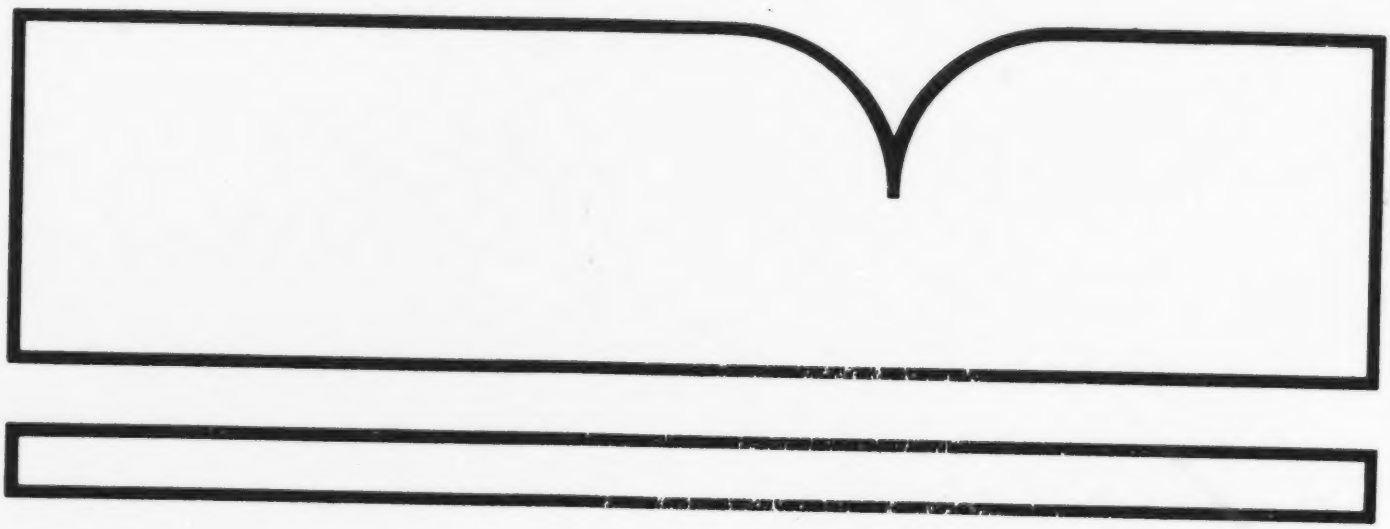

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SELENIUM REMOVAL USING THE CONVENTIONAL COAGULATION OR LIME-SODA ASH SOFTENING PROCESS

by

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16. ABSTRACT

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ABSTRACT

To develop treatment technology information in support of the USEPA selenium drinking water regulation of 0.01 mg/L, the Drinking Water Research Division, USEPA, Cincinnati, OH, conducted laboratory jar tests and pilot plant tests for selenium IV and VI removal by conventional coagulation and lime-soda ash softening. This paper presents the results of these studies that showed removals to be dependent on the valence and concentration of selenium, type and dose of coagulant, and pH of the feed water. Selenium VI was poorly removed by both processes while selenium IV was moderately removed.

INTRODUCTION

The U.S. Environmental Protection Agency (USEPA) National Interim Primary Drinking Water Regulations (NIPDWR) (1) and the 1984 World Health Organization (WHO) guidelines (2) for drinking water have established the maximum level for selenium in drinking water at 0.01 mg/L. Presently the USEPA is in the process of issuing its final drinking water regulations. Based upon a review of the most recent health information, current estimates of selenium intake from other sources, and the fact that current data suggests that selenium is an essential nutrient and has anti-carcinogenic effects in animals, preliminary indications are that USEPA will propose to raise the MCL by a factor of 3 or 4 (0.03 - 0.04 mg/L).

Because selenium is not commonly found in high concentrations in either drinking water or wastewater, treatment information is not readily available in the literature. To support the NIPDWR promulgated in 1975, the Drinking Water Research Division (DWRD), Office of Research and Development, USEPA Cincinnati conducted laboratory and pilot plant studies in the 1970's to evaluate various unit treatment processes for the removal of selenium including conventional coagulation and lime softening (3). Since that time, EPA has funded several other projects on the evaluation of activated alumina (4,5), ion exchange (6,7) and reverse osmosis (8). The results of several of these projects are being reported upon by other investigators at this seminar. The objective of this paper is to review the results of laboratory and pilot plant studies on our conventional coagulation and lime softening studies for selenium removal.

SELENIUM CHEMISTRY OVERVIEW

Selenium has several oxidation states, but only two are predominant in water: selenium IV (selenite) and selenium VI (selenate). Both forms

occur as anions and have acid characteristics.

Selenite (Se IV) in the form of selenious acid (H_2SeO_3) is a weak acid. Selenious acid will dissociate to $HSeO_3^-$, which predominates in the pH range of 2.7 to 8.5, and to $SeO_3^=$, which prodominates above pH 8.5. Thus, the major selenite specie associated with drinking water is the monovalent $HSeO_3^-$.

Selenate (Se VI) in the form of selenic acid (H_2SeO_4) is comparable in strength to sulfuric acid. Above pH 1.7, the major specie is $SeO_4^=$, a divalent ion that is chemically similar to sulfate.

The chemistry of selenite and selenate shows that selenate salts are generally much more soluble than the corresponding selenite salts. Thus, selenite was anticipated to have higher removals than selenate by both conventional coagulation and lime softening. Because selenate is similar to sulfate, both processes were expected to be ineffective for selenate removal.

Although the literature reveals that little data and information exists on the removal of selenium by conventional water treatment and lime softening processes, several chemistry studies have been conducted on the adsorption of selenium by ferric hydroxide (9-14). Although the results do not apply directly to water treatment, the work is related and the results provide some insight to the potential results of water treatment processes. Most of the significant information from these studies showed that selenite is more readily adsorbed than selenate and that selenite adsorption is pH dependent. The most effective pH range is below pH 8: above pH 8, selenite adsorption drops off very sharply.

Two wastewater studies have been reported upon on the removal of selenite (15-16). Linstedt et al. investigated the removal of selenite in a pilot plant consisting of coagulation, sedimentation, sand filtration, activated carbon and ion exchange (15). The wastewater was secondary effluent from a wastewater treatment plant having trickling filters. The natural selenium concentration was less than 0.002 mg/L but increased to 0.013 mg/L with a selenite spike. Treatment with lime to pH 11 and sedimentation achieved 16.2 percent removal. Sand filtration alone produced 9.5 percent removal.

Hannah et al. reported on the removal of selenite from raw sewage in three physical-chemical pilot plants (16). The treatment train for the three plants consisted of coagulation, sedimentation, dual-media filtration, and carbon filtration. Two activated carbon filters were used in parallel following the dual media filter. The three plants were run simultaneously using different coagulants; (1) alum, (2) ferric sulfate, and (3) lime. The raw sewage was spiked with selenite (0.06 - 0.5 mg/L Se IV). Iron coagulation at pH 6-7 achieved the highest removals, 75-80 percent with 0.05 - 0.1 mg/L selenium IV. Alum coagulation (pH 6-7) removal of 0.5 mg/L selenium IV was lower, 50 percent, and lime coagulation, the least, 35 percent, with 0.5 mg/L of selenium IV.

SELENIUM STUDIES

Initially, laboratory jar tests studies were conducted to evaluate the many variables associated with the conventional coagulation and lime softening processes. These tests were conducted using spiked groundwater (Glendale, OH) and surface water (Ohio River Water) obtained in the

Cincinnati area. Both valences of selenium (IV and VI) were investigated. The variables studies for the conventional coagulation jar tests included pH of the source water, coagulant (alum, ferric sulfate and ferrous sulfate), coagulant dose, and selenium concentration. In the lime softening tests, the effect of pH of the water (lime dose) and selenium concentration were studied. Once optimum treatment conditions were determined, pilot plant studies were conducted with the same two source waters and a gravel pit water using a 2 gpm conventional coagulation/filtration pilot plant (3). This same plant was also used for the lime softening pilot plant studies.

Because of difference in the water matrices, the results of the USEPA studies for drinking water purposes, may not necessarily extrapolate to other waters, particularly those associated with the San Louis drainage waters. The major differences in the types of waters is the high TDS level of the San Louis drainage waters (8,000-10,000 mg/L). The main constituents of these waters are sulfate, sodium, chloride, and hardness (Ca and Mg). The selenium concentration of 3-4 mg/L of the drain water is also considerably higher than that normally found in drinking water sources (0.05 - 0.5 mg/L). The results of this work, however, should give some general indication of the potential application for the processes studied to the San Louis drainage water.

RESULTS

The results of jar tests and pilot plant studies on selenium IV and VI showed a significant difference in removal between the two valences by both processes reported on; conventional coagulation and lime softening.

Selenate Removal

Because selenate is similar to sulfate, low removals by both conventional coagulation and lime softening were anticipated and the results of the tests confirmed it. Conventional coagulation studies with iron (ferric sulfate) and alum from pH 6 - 8 and with coagulant doses as high as 200 mg/L achieved less than 10 percent removal of selenate under all conditions in both groundwater and surface water (Table 1).

Jar tests were also conducted with ferrous sulfate (20 - 100 mg/L) as a coagulant because of its reducing capability, but no significant increase in removals were achieved (Table 1). Powdered activated carbon (5 - 100 mg/L) was also tried with no success (less than one percent removal).

Lime softening experiments were conducted on a hard ground water with results similar to conventional coagulation; removal of selenate (0.03 - 0.1 mg/L) was less than 10 percent through the pH range of 9.0 - 11.5 (Table 2).

Pilot-plants tests were conducted on low levels of selenate by conventional coagulation and lime softening with the results being similar to the jar tests results. Less than 10 percent removal was obtained by both processes under all conditions studied (Table 3).

Selenium IV Removal

Jar tests and pilot plant tests conducted on the removal of selenium IV from well water and surface waters by conventional coagulation showed that

Table 1. Results of Jar Test Studies on Selenium VI Removal by Conventional Coagulation

Coagulant-dose mg/L	Water Source	Se VI Conc. of Raw Water - mg/L	pH of Water	Percent Removal
Ferric Sulfate	Ground			
25		0.1	6.4	3
60		0.1	6.3	7
100		0.1	6.3	4
Ferric Sulfate	River			
25		0.1	7.4	9
60		0.1	7.3	4
100		0.1	7.2	5
Ferric Sulfate	Ground			
25		0.1	8.2	1
60		0.1	8.1	4
100		0.1	7.9	2
Alum	Ground			
25		0.1	6.0	0
60		0.1	5.8	2
100		0.1	5.7	5
Alum	River			
25		0.1	7.5	0
60		0.1	7.3	2
100		0.1	6.9	3
Alum	Ground			
25		0.1	8.3	1
60		0.1	8.1	1
100		0.1	7.9	2
Ferric Sulfate	Ground			
40		0.03	6.3	2
100		0.03	6.1	2
200		0.03	5.8	7
Alum	Ground			
40		0.03	6.4	0
100		0.03	6.2	2
200		0.03	6.0	5
Ferrous Sulfate	River			
10		0.03	9.0	2
20		0.03	9.0	8
30		0.03	9.0	9
50		0.03	9.0	10
75		0.03	9.2	3
100		0.03	9.1	4
Ferrous Sulfate	Ground			
20		0.1	7.4	3
20		0.1	9.5	0
20		0.1	10.2	1
20		0.1	7.9	0
20		0.1	9.5	0
20		0.1	10.3	2

Table 2. Results of Jar Test Studies on Selenium VI Removal by Lime Softening Treatment

Lime-dose mg/L	Water Source	Se VI Conc. of Raw Water - mg/L	pH of Water	Percent Removal
Lime	Ground			
200		.03	8.9	2
325		.03	10.3	1
475		.03	10.8	2
Lime	Ground			
200		0.1	9.4	3
325		0.1	10.3	1
475		0.1	10.9	3

Table 3. Results of Pilot Plant Tests on Selenium VI Removal by Conventional Coagulation and Lime Softening Treatment

Coagulant Dose mg/L	Source Water	Avg. Se VI Concentration mg/L	pH of Treated Water	Percent Removal	
				Settled Water	Filtered Water
Ferric Sulfate-32	River	0.097	6.5	11	14
Alum - 29	River	0.076	8.1	6	8
Alum - 30	River	0.055	8.3	0	5
Ferrous Sulfate-29	River	0.042	8.1	0	0
Ferrous Sulfate-32	River	0.083	8.3	1	2
Alum - 25	River	0.028	6.8	18	29
Lime	Ground	0.038	9.5	11	3
Lime	Ground	0.038	11.3	16	8

selenium IV removal is dependent on the pH of the treated water, the coagulant and coagulant dose, and the raw water selenium concentration. The results of the jar tests to evaluate pH and coagulant type are shown in Figure 1. The removal curves show that iron coagulation (ferric sulfate) is much more effective than alum coagulation and that iron coagulation is very pH dependent. The highest removals (~80%) achieved were with ferric sulfate in the pH range 6-7 with Ohio River water. Removals decreased to a minimum near 10 percent at pH 9.

Alum coagulation was found to be independent of pH and removals were significantly less (10 - 25 percent) than iron coagulation. Figure 1 also shows that removals by both coagulants were about 5 - 10 percent less with the well water than with river water in the pH range 6 - 8.5. Above pH 8.5, removal by both coagulants with both types of water were almost identical, at 15 - 20 percent removal.

Higher selenium IV removals can be achieved by increasing the dose of either coagulant (Figure 2). The improvements achieved by increasing the coagulant dose at low pH are significantly smaller than those for pH 8.5 because the initial removals with low doses (25 mg/L) are much higher in the low pH range. Even though there was little pH dependency, alum coagulation with higher doses produced the greater overall increases in removals in the lower pH range.

Whereas the initial pH evaluation jar tests were run with a selenium IV concentration of 0.03 mg/L, the dose response tests were conducted with a concentration of about 0.1 mg/L. These concentrations are representative of concentrations that might be found in water supply, but significantly lower than the 3 - 4 mg/L selenium concentration of the San Louis drain water. Jar tests were conducted with higher concentration (0.1 - 10 mg/L) to determine the percentage removal dependency on initial concentration and the results are shown in Figure 3. Once again the readers are reminded that the water sources for these tests were significantly different than the San Louis drain water. With iron coagulation (25 mg/L) at pH 7.2, removals decreased from 58 percent to near 30 percent as the selenium concentration increased from 0.1 to 10 mg/L. With 3 mg/L Se IV, removals were 38 percent resulting in a treated water concentration of 1.86 mg/L, which is far above the drinking water MCL. Because alum is less effective than iron, results of these tests were even less favorable. With 25 mg/L alum at pH 7.7 only about 5 percent removal was achieved with 3 mg/L selenium IV in groundwater.

Because selenate is very poorly removed by conventional coagulation, and selenite can be oxidized to selenate, several jar tests were conducted to determine whether prechlorination would have an impact on selenium IV removal. Tests were conducted on spiked well water having a low chlorine demand (0.2 - 0.5 mg/L). The tests results at several pH levels (6.4 to 8.1) as shown in Table 4 indicate that chlorination could lower selenium removals by converting the selenite to selenate. The test with 2 mg/L at pH 6.4 showed a reduction in removal from 56 to 21 percent removal with 60 minutes contact time. Removal decreases were also found with tests run at pH 7.9, 8.0 and 8.1. Several tests (pH 6.8, 7.4, 7.8), however did not show a decrease. Why no reductions occurred during these tests is not known. Furthermore, the results are not completely consistent with the results of the oxidation studies of Baegel and Clifford (7). These investigators showed that the best pH range for selenium IV oxidation by chlorine (2 mg/L) was pH 6.5 - 7.5. Above and below this range, the percent of oxidation decreased (Figure 4). The results of the jar tests coupled with the fact that selenite can oxidize to selente, suggest that preoxidation with chlorine should not be practiced to obtain best removals of selenite by either conventional coagulation or lime softening.

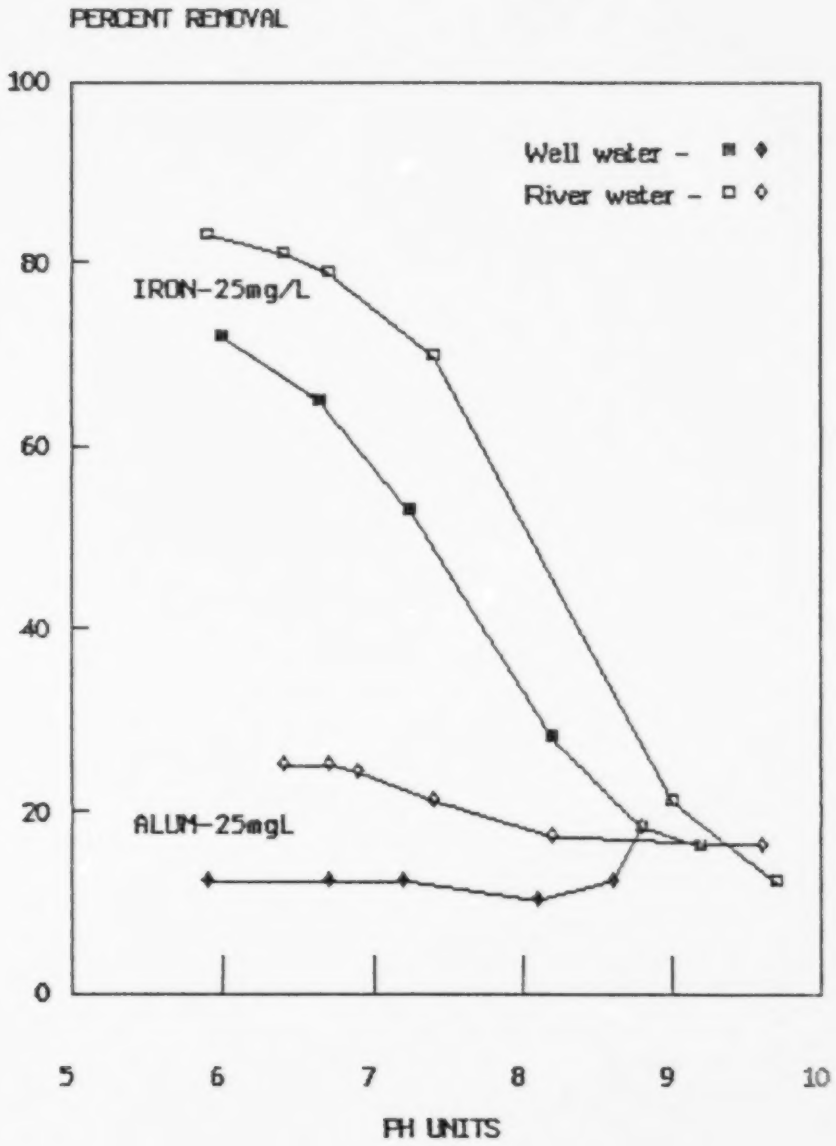

Figure 1 Effect of pH on Removal of Selenium IV (0.03 mg/l) by Conventional Coagulation Treatment.

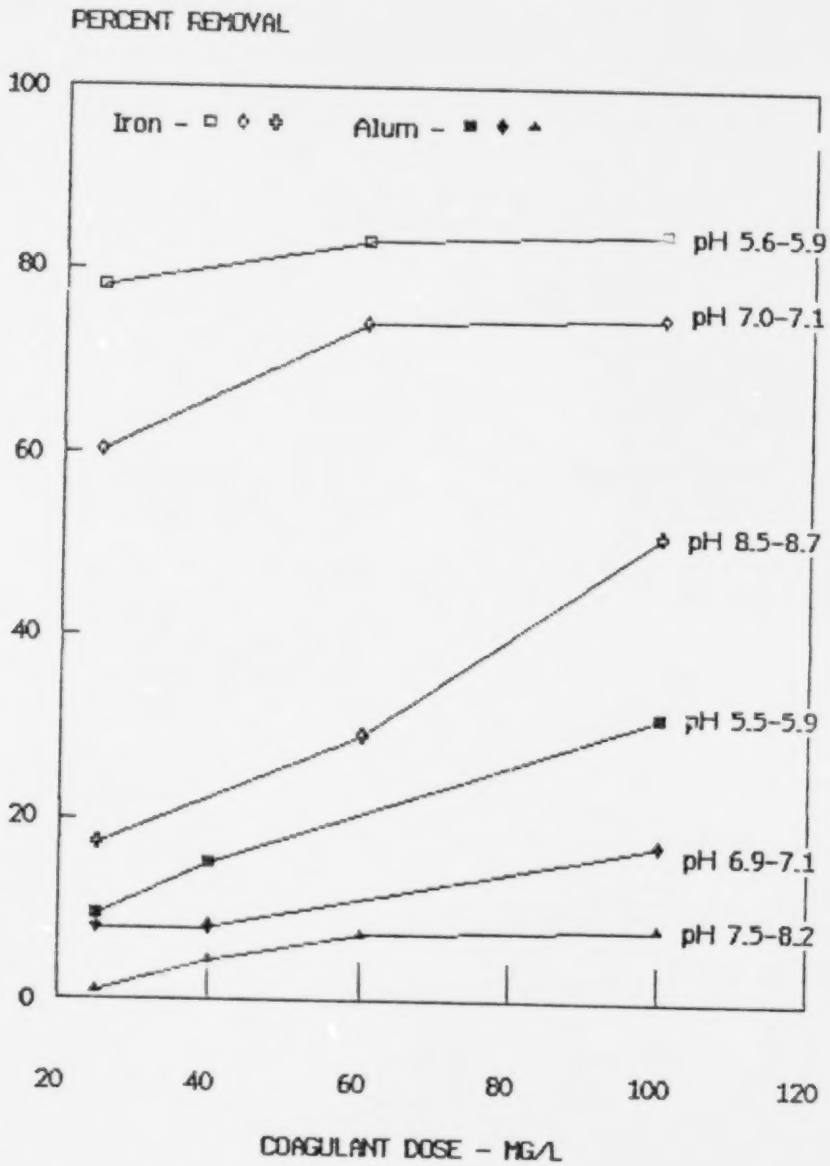

Figure 2 Effect of Coagulant Dose on Selenium (0.1 mg/L) Removal From Ground Water by Conventional Coagulation Treatment.

Table 4. Effect of Prechlorination on Selenium IV Removals by Iron Coagulation (25 mg/L).

pH of Ground Water Units	Chlorine Dose mg/L	Contact Time* Minutes	Percent SeIV Removal**
7.6	0	0	25
7.7	1	5	19
7.6	1	10	18
7.6	1	20	18
7.7	1	60	19
6.8	0	0	31
6.8	1	5	31
6.7	1	10	28
6.7	1	20	29
6.8	1	60	29
8.1	0	0	30
8.1	2	5	5
8.1	2	10	4
8.1	2	20	2
8.2	2	60	0
8.0	0	0	29
8.0	2	5	1
8.0	2	10	3
8.0	2	20	1
8.0	2	60	1
7.9	0	0	28
7.8	2	5	6
7.8	2	10	4
7.8	2	20	2
7.8	2	60	0
7.8	0	0	30
7.7	2	5	27
7.7	2	10	29
7.7	2	20	27
7.7	2	60	28
7.5	0	0	39
7.4	2	5	37
7.4	2	10	38
7.4	2	20	37
7.4	2	60	37
6.4	0	0	56
6.4	2	5	45
6.4	2	10	43
6.4	2	20	36
6.4	2	60	21

* Chlorine measured at test completion and residual of 0.5+ mg/L was always present.
**Initial selenium concentration of ground water was 0.1 mg/L.

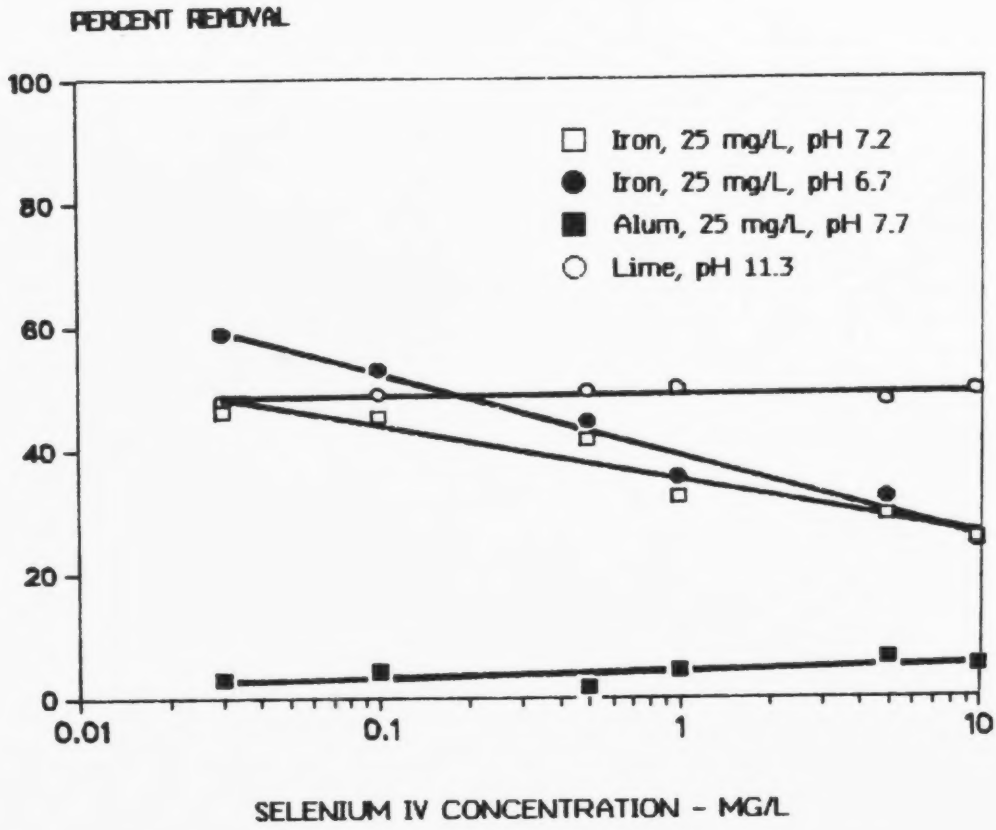

Figure 3 Effect of Initial Concentration on Selenium IV Removal by Conventional Coagulation and Lime Softening Treatments.

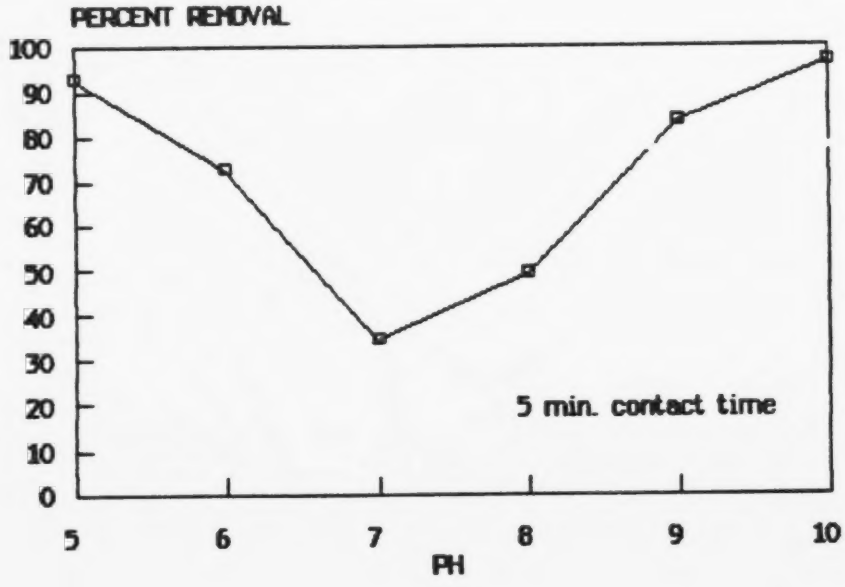

Figure 4 Effect of pH on Oxidation of Selenium IV with Chlorine.

Pilot plant tests for selenium IV removal were conducted as a follow up to the jar test studies using the DWRD 2 gpm pilot plant system. The water sources (spiked) for these tests were two surface waters, Ohio River water and a Cincinnati area low turbidity gravel pit water, and Glendale, Ohio well water. The results of 13 test runs are given in Table 5. The results parallel the jar test studies. Ferric sulfate was more effective than alum and highest removals were achieved at the low pH of 6.5. Furthermore, slightly higher removals were generally obtained with river water than with the low turbidity (0.05 - .5 ntu) well water.

Jar test studies on the removal of selenium IV by lime softening showed that this treatment method is only moderately effective. Test results (Figure 5) were pH dependent with maximum removal of selenium IV (0.1 mg/L) being about 50 percent at pH 11.5. Below this pH, removals decreased with decreasing pH. Jar test studies to evaluate the effect of the initial selenium concentration were conducted at pH 11.5. In the range of 0.05 to 10 mg/L selenium IV range, removals remained constant at about 50 percent removal (Figure 3). Thus, as the initial concentration increases, the amount of selenium IV remaining in the treated water increases.

To confirm the jar test work, two pilot plant test runs were conducted at pH 9.5 and 11.3. The test results, as given in Table 5, were similar to the jar test results; 30 percent at pH 9.5 and 46 percent at 11.3.

SUMMARY AND CONCLUSIONS

The removal of selenium from water and wastewater has not been thoroughly investigated on a full-scale treatment plant basis. Laboratory and pilot plant studies have been conducted, however, and the results indicate that conventional coagulation and lime softening treatment process methods are ineffective for selenate removal and only moderately effective for selenite removal

The general concluions drawn from the drinking water and wastewater studies are as follows:

1. Conventional water treatment techniques are ineffective for selenate removal; no more than 10 percent is achieved by alum or iron coagulation, or lime softening.

2. Iron coagulation is the most effective conventional water treatment method for the removal of selenite from water (60-80 percent). Iron coagulation is also pH dependent; removals increase as the pH of the treated water decreases with best results at pH less than 7.

3. Alum coagulation and lime softening are less effective than iron coagulation for selenite removal. Lime softening can achieve 20 - 40 percent removal with removals increasing with pH increase from pH 9 to 11. Alum is the least effective with only 10 - 20 removal efficiency.

4. Selenium IV can be oxidized to selenium VI by chlorine and thus prechlorination will decrease the removal of selenite.

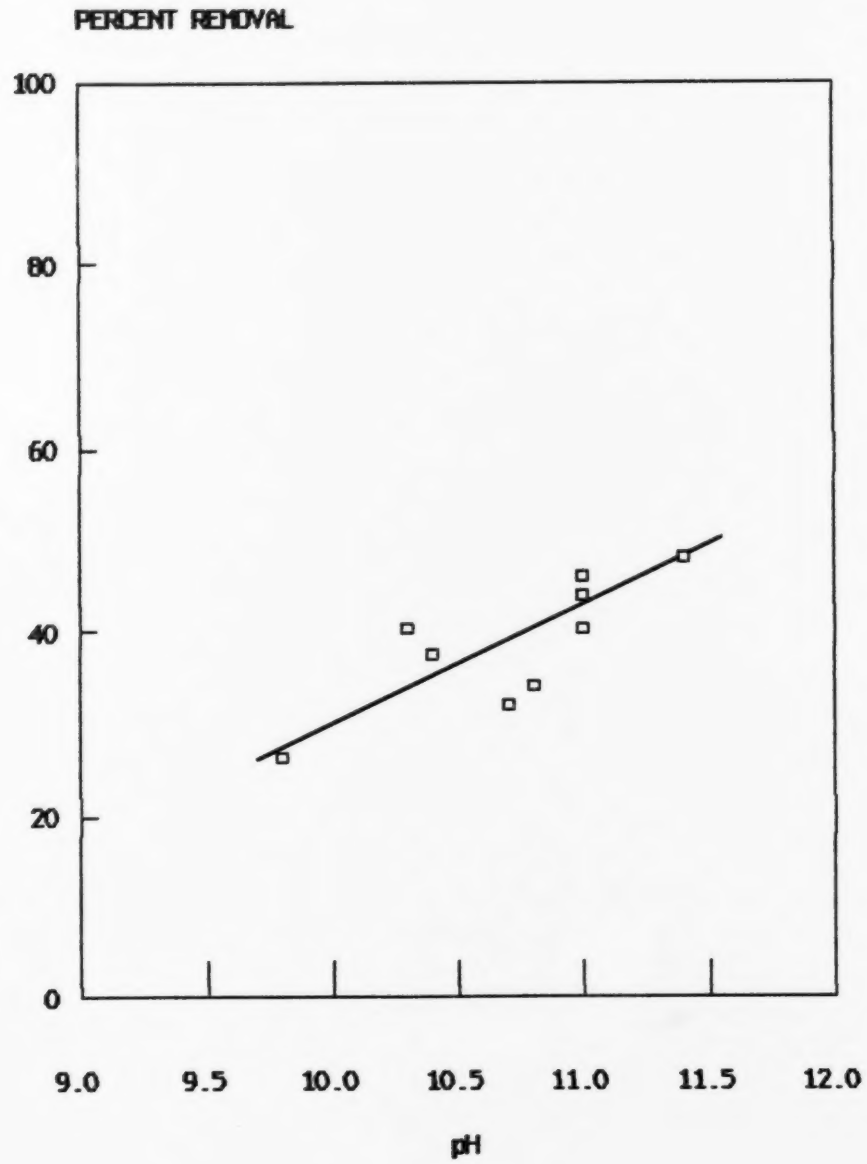

Figure 5 Effect of pH on Selenium IV (0.1 mg/L) Removal by Lime Softening.

Table 5. Results of Pilot Plant Tests on Selenium IV Removal by Conventional Coagulation and Lime Softening Treatment

Coagulant Dose mg/L	Source Water	Avg. Se IV Concentration mg/L	pH of Treated Water	Percent Removal	
				Settled Water	Filtered Water
Ferric Sulfate-15	Gravel Pit	0.04	6.5	20	58
Ferric Sulfate-15	Gravel Pit	0.05	8.5	0	0
Alum - 27	Gravel Pit	0.061	8.5	5	8
Alum - 20	Gravel Pit	0.066	7.5	23	20
Alum - 17	Gravel Pit	0.050	7.5	4	10
Alum - 15	Gravel Pit	0.033	6.5	3	6
Ferric Sulfate-25	River	0.017	8.3	26	30
Ferric Sulfate-25	River	0.025	7.8	64	78
Ferric Sulfate-23	River	0.027	6.9	81	81
Alum - 31	River	0.019	6.6	12	20
Alum - 30	River	0.029	8.3	29	20
Alum - 34	River	0.022	7.9	14	18
Ferric Sulfate-29	River	0.024	7.9	47	48
Alum - 28	Ground	0.056	7.8	18	29
Ferric Sulfate-33	Ground	0.079	7.5	69	74
Ferric Sulfate-30	Ground	0.047	6.4	53	79
Lime	Ground	0.038	9.5	21	30
Lime	Ground	0.028	11.3	38	46

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